

Method of reducing the emission of nitrogen oxides from coke ovens

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Please cite as: CHEMIK 2016, 70, 10, 597–604

Introduction

The production of coke in coke oven batteries is related to the emission of substances into the air, including the emission of nitrogen oxides [1]. Nitrogen oxides may constitute 16 to 41% of the total emission of substances from coke oven batteries [2].

The combustion of coke gas in the battery heating system called the coke-oven battery firing is the main source of the emission of nitrogen oxides NO_x from the coke-oven battery [1].

Carbonization of hard coal consists in its degassing by preventing the air flow into the coke ovens at a temperature of ca. K. The coke oven consists of a horizontal coking chamber, indirectly heated by combusting coke oven gas in the heating walls. A set of alternately situated coking chambers and heating walls forming a compact unit of ovens is called the coke oven battery [6].

In Poland 25 coke oven batteries are operated with a production capacity of 11 million tonnes of coke per year. In the years 1998–2012 the coke oven batteries were technologically modernized in order to reduce emissions [4]. Ten new coke oven batteries were built by using modern carbonization techniques and environmental protection. The remaining 15 coke oven batteries are successively modernized and equipped with devices for air tight sealing or restricting emissions to the air [5].

Environmental requirements for the years 2012–2020 for coking installations were defined in the Directive 2010/75/EU of the European Parliament and of the Council of 24th November 2010 on industrial emissions and in the Commission Implementing Decision of 28th February 2012 establishing BAT conclusions for the iron and steel production [7, 8].

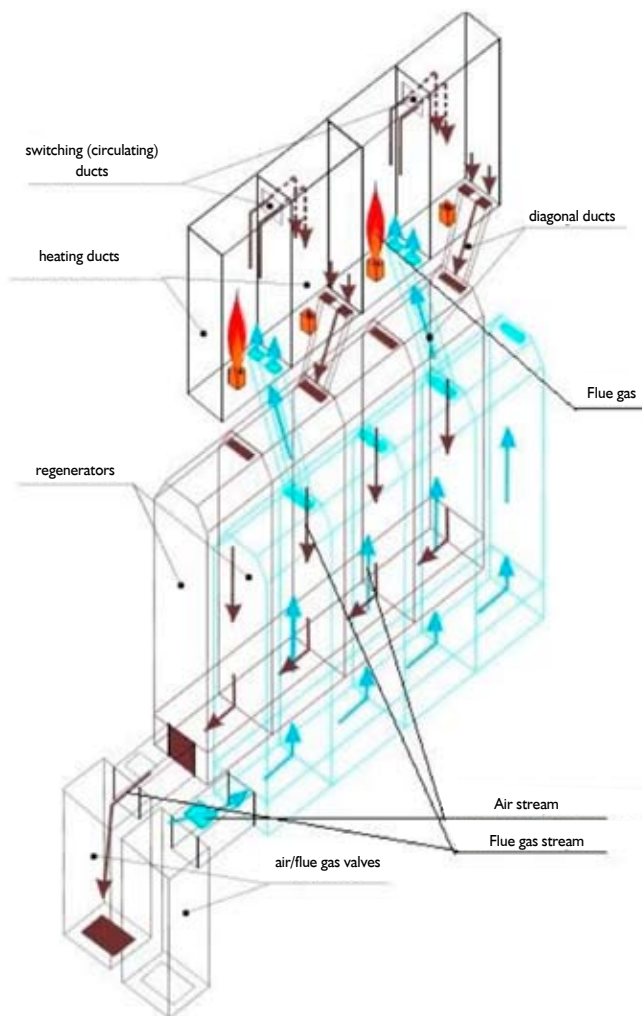
The object of work is the analysis of the effectiveness and availability of methods of reducing the emission of nitrogen oxides from coke oven batteries and, in particular, from the process of combusting coke oven gas in the battery heating system.

Formation and reduction methods of the emission of nitrogen oxides in the heating system of the coke oven batteries

The carbonization of coal requires the supply of a large amount of heat which is obtained by combusting coke oven gas in the coke oven battery heating system.

The coke oven battery heating system is comprised of heating walls which are split into twin heating ducts with burners and air-flue gas inlets (outlets) operating with regenerators, heating and changeover accessories and linked through smoke ducts and the stack. In the twin heating ducts gas is alternately combusted in one duct and flue gases are discharged through the other one [9, 10]. The hydraulic diagram of the coke oven battery heating system was presented in Illustration 1 [11].

The combustion of coke oven gas in the coke oven battery heating system is related to the formation and emission of NO_x [2].



Explanation:

- Air stream
- Flue gas stream

Illustration 1. Hydraulic diagram of the heating system of coke oven batteries [11]

Three mechanisms can be distinguished in the formation of NO_x : the thermal, fast and fuel (slow) mechanism [12]. The thermal mechanism of forming nitrogen oxides is based on oxidation reactions of nitrogen from air used for combusting coke oven gas. This mechanism is crucial when the flame temperature is higher than 1673 K. The key factor for the reduction of thermal nitrogen oxides in flue gases is the flame temperature decrease and the reduction of time and concentration of N_2 and O_2 in the high temperature zone [13].

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Fast nitrogen oxides may form during combustion of methane contained in coke oven gas in the subflame zone where the temperature does not exceed 1073 K. The amount of produced NO_x depends on the concentration of molecular nitrogen in the sub-flame zone [12].

In coke oven gas, there is a minimum percentage of chemically bonded nitrogen. It may be assumed that during the combustion of coke oven gas, the fuel nitrogen oxides are negligible [2]. A series of methods of reducing the NO_x emission was worked out and implemented in the new and modernized coke oven batteries. These methods reduce the formation of NO_x at the source itself, ie. during the combustion of coke oven gas in the heating ducts; these are the original methods of reducing the NO_x emissions [11]. These methods include: reduction of the gas combustion temperature, internal and external flue-gas recirculation to heating ducts and gradual combustion via a multistage injection of air or gas to the heating duct [14 ÷ 16].

The implementation of these methods is possible at the construction stage of new coke oven batteries, what results from the necessity to introduce design changes in the ceramic mass of the coke ovens [17].

Secondary measures, ie. fuel denitration are not used to reduce the emission of nitrogen oxides from the combustion of coke oven gas in the heating system of the coke oven batteries. Industrial research in Japan demonstrated that these measures have no technical and economic justification [3, 19].

In the years 1998 until 2012 ten new coke oven batteries were built in which effective NO_x emission reduction methods were used [5]. The NO_x emission from the combustion from the new coke oven batteries dropped from 820 mg of NO₂/m³_u to 490 mg of NO₂/m³_u of flue gases calculated as 5% of the oxygen content in flue gases [18].

Methods of reducing emissions of nitrogen oxides from the existing coke oven batteries

The implementation of primary methods of reducing the emissions of nitrogen oxides at industrial scale is related to design changes on the coke oven battery. This is possible when building new coke oven batteries [17, 19]. The emission of nitrogen oxides in the battery heating system can be reduced in the existing coke oven batteries via: lowering the temperature of carbonization of coal, aromatization of coke oven gas and optimization of hydraulic-temperature parameters of the coke oven battery heating system [2, 20].

The reduction of the carbonization temperature in the coke ovens lowers NO_x emission, but results in prolonging carbonization time and a lower production capacity utilization of the batteries [21].

The impact of the coal carbonization temperature in coke ovens on the concentration of nitrogen oxides in flue gases was presented in Table I.

Table I
Impact of the coal carbonization temperature on the concentration of nitrogen oxides in flue gases [22]

| Item No. | Description | Unit | Value | | | |
|----------|--|---|------------------------|-----------------------|------------------------|-----------------------|
| | | | PTU-57 batteries | | PWR-63 batteries | |
| | | | State before reduction | State after reduction | State before reduction | State after reduction |
| 1 | 2 | 3 | 4 | 5 | 6 | 7 |
| 1 | Carbonization temperature | °C | 1320 | 1260 | 1330 | 1260 |
| 2 | Carbonization time | h | 20 | 26 | 16 | 22 |
| 3 | Concentration of nitrogen oxides in flue gases | Mg NO ₂ /m ³ _u (5 % O ₂) | 986 | 584 | 618 | 382 |

An effective way of reducing the concentrations of nitrogen oxides in flue gases in the existing coke oven batteries is the increase of the content of aromatic hydrocarbons in coke oven gas [2, 23, 24]. Coke oven gas combusted in the coke oven battery heating system usually contains aromatic hydrocarbons (benzene, toluene, xylene) up to 3 g BTX/m³_u of gas. The increase of aromatic hydrocarbons in the coke oven gas combusted in the battery heating system from 3 g BTX/m³_u to 12 g BTX/m³_u reduces nitrogen oxides in flue gases.

This is demonstrated in Table 2 worked out on the basis of testing of the emission value of nitrogen oxides from firing the PTU-57 coke oven battery depending on the content of aromatic hydrocarbons in the coke oven gas [23, 24]:

Table 2

| Item No. | Description | Unit | Value | | | |
|----------|--|---|-------|------|------|------|
| | | | 4 | 5 | 6 | 7 |
| 1 | Carbonization temperature | °C | 1320 | 1320 | 1320 | 1320 |
| 2 | Concentration of BTX hydrocarbons in gas | g/BTX/m ³ _u | 2.9 | 6.1 | 9.2 | 11.9 |
| 3 | Concentration of oxygen in flue gases | % O ₂ | 9.8 | 9.6 | 9.7 | 9.9 |
| 4 | Concentration of nitrogen oxides in flue gases | Mg NO ₂ /m ³ _u (5 % O ₂) | 994 | 724 | 582 | 466 |

The increase of the content of aromatic hydrocarbons in coke oven gas effectively reduces the emission of nitrogen oxides from firing coke oven batteries. It should be emphasized that a higher concentration of aromatic hydrocarbons in coke oven gas increases the calorific value of gas what allows to obtain additional energy effects.

The implementation of the method of reducing the concentrations of nitrogen oxides in flue gases on coke oven batteries via increasing the aromatization of coke oven gas should be preceded by industrial research in terms of safety and process efficiency on a given coke oven battery [3, 10, 19].

The third method of reducing the emissions of nitrogen oxides from the existing coke oven batteries is the optimization of the process of firing batteries consisting in: changing the way of providing coke oven gas to the battery heating system – from continuous to periodic provision and normalization of hydraulic-temperature parameters of the battery heating system. The concentration of nitrogen oxides dropped on the PTU-57 battery from 23% to 37% in relation to the concentration of nitrogen oxides in flue gases prior to the application of these methods [11, 25, 26].

The assessment of the possibility to meet the emission requirements for coke oven batteries

The environmental requirements in the scope of the emission of nitrogen oxides from coke oven batteries were defined in the Commission Implementing Decision (of 28th February 2012) establishing conclusions concerning the Best Available Technology (BAT) in accordance with the Directive 2010/75/EU of the European Parliament and of the Council on industrial emissions in relation to the iron and steel production [8].

The BAT conclusion for the coke oven batteries defines the admissible emission standards for nitrogen oxides:

- <350–500 mg NO₂/N_u³ in relation to 5% of O₂ in flue gases for new or substantially modernized coke oven batteries,
- <500–650 mg NO₂/N_u³ in relation to 5% of O₂ in flue gases for existing coke oven batteries suitably operated with the use of technical solutions for reducing nitrogen oxides (NO_x) emission.

In the years 1998 – 2012 ten new coke oven batteries were built in which the effective NO_x emission reduction methods were used [5]. The NO_x emission from firing the new coke oven batteries dropped from $820 \text{ mg NO}_2/\text{m}^3_u$ to $490 \text{ mg NO}_2/\text{m}^3_u$ of flue gases as calculated on 5% oxygen content in flue gases [18].

The NO_x emission standards for these batteries are maintained.

Programmes adapting the coke oven batteries to the emission requirements of nitrogen oxides resulting from BAT conclusion are carried out for the existing systems.

Among the most important actions are [5, 20]:

- renovation of the ceramic mass and of the system for collecting raw gas from the battery and coke oven gas cleaning,
- implementation of effective NO_x reaction methods.

Summary

Nitrogen oxides (NO_x) are the crucial air pollutants from coke oven batteries. The percentage of the emission of nitrogen oxides in the total emission from the coke oven battery varies from 16% to 41% [1].

The primary source of the emission of nitrogen oxides is the combustion of coke oven gas in the battery heating system, i.e. battery firing. The emission of nitrogen oxides (converted into NO_2) from the aforementioned source varies from 380 to 1350 g/Mg of coke [1]. Heating the coke ovens (battery firing) consists in bringing coke oven gas via distribution pipes to burner channels and their corresponding heating ducts. In the heating ducts gas is burnt in the excess of air supplied through the intake holes from air and flue gas switching valves and regenerators. Flue gases from the fired heating ducts flow through the circulating port to the adjacent ducts and then they are extracted via ducts through the distribution grate to the regenerators, air and flue gas switching valves, flue gas collectors and the stack [9, 10].

The dominant mechanism of the formation of nitrogen oxides during gas combustion in the battery heating system is the oxidation of nitrogen contained in air via the oxygen radical formed in the reaction zone at a temperature above 1673 K [12]. Solutions reducing the formation of nitrogen oxides are used to decrease the emission of nitrogen oxides from new coke oven batteries. They are based on design changes of the ceramic mass of the battery what allows reducing the gas combustion temperature, internal and external recirculation of flue gases to heating ducts and gradual gas combustion through a multi-stage introduction of air or gas into the heating duct [14 ÷ 16].

In the existing coke oven batteries it is possible to reduce the emission of nitrogen oxides in the battery heating system via: lowering the coal carbonization temperature, aromatization of coke oven gas and the optimization of hydraulic-temperature parameters of the coke oven battery heating system [24 ÷ 26].

Environmental requirements in the scope of the emission of nitrogen oxides from coke oven batteries were defined in the BAT conclusion and, in particular, admissible emission standards from the battery firing were determined [8]:

- $<350\text{--}500 \text{ mg NO}_2/\text{N}_u^3$ in relation to 5% of O_2 in flue gases for new or substantially modernized coke oven batteries,
- $<500\text{--}650 \text{ mg NO}_2/\text{N}_u^3$ in relation to 5% of O_2 in flue gases for existing coke oven batteries suitably operated with the use of technical solutions for reducing nitrogen oxides (NO_x) emission.

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- implementation of effective NO_x reaction methods.

The presented analysis of the methods of reducing nitrogen oxides from firing coke oven batteries worked out and implemented in industrial practices and also the adjustment of programmes to the requirements of BAT conclusion by the operators should enable the coke oven batteries to meet the environmental requirements. The older and used coke oven batteries are problematic because the possibilities for reducing NO_x from battery firing are limited.

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Lanxess przejmie producenta dodatków chemicznych

Koncern Lanxess ogłosił plan przejęcia amerykańskiej firmy Chemtura Corporation, będącej jednym z głównych dostawców wysokiej jakości dodatków opóźniających palenie i smarów. To największe w historii koncernu Lanxess przejęcie pomoże mu poszerzyć swoje portfolio dodatków do tworzyw sztucznych i stać się jednym z najważniejszych na świecie podmiotów działających na tym ciągle rozwijającym się rynku. Firmy podpisały ostateczną umowę o przejęciu, zgodnie którą akcjonariusze firmy Chemtura otrzymają 33,50 USD w gotówce za każdą jej akcję zwykłą pozostającą w obrocie, co stanowi 18,9% nadwyżkę względem ceny zamknięcia z dnia 23 września 2016 r., która wyniosła 28,18 USD. Przedmiotowa transakcja, która dotyczy przedsiębiorstwa o wartości ok. 2,4 mld EUR, zostanie sfinansowana przez koncern Lanxess głównie za pomocą emisji obligacji z prawem pierwszeństwa oraz obligacji hybrydowych, a także z dostępnych środków finansowych. Zamknięcie transakcji powinno nastąpić mniej więcej w połowie 2017 r., po jej zatwierdzeniu przez akcjonariuszy firmy Chemtura, uzyskaniu odpowiednich zgód od organów regulacyjnych oraz spełnieniu innych zycząjowych warunków realizacji tego typu przedsięwzięć. (kk)

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NOWE INWESTYCJE

Polski producent poliuretanów przymierza się do inwestycji w Tajlandii

Zarząd PCC Rokita poinformował o dalszych ustaleniach w sprawie rozważanej możliwości rozpoczęcia w Tajlandii produkcji w branży poliuretanowej. Polska spółka dokonała analizy sytuacji oraz potencjału istniejącej spółki IRPC Polyol Company Ltd. w Bangkoku, wchodzącej w skład grupy IRPC, jako producenta systemów poliuretanowych i polioli. Analiza miała na celu zbadanie możliwości inwestycji w IRPC Polyol poprzez zakup części udziałów w tej spółce i utworzenie spółki joint venture wspólnie z IRPC Public Co. Ltd. w Bangkoku. Zarząd PCC Rokita pozytywnie ocenił zasadność inwestycji i zwrócił się do rady nadzorczej spółki o wyrażenie stosownych zgód na inwestycję.

W ramach prowadzonych negocjacji z IRPC Public Co. Ltd. w Bangkoku, ustalona została cena nabycia od tej spółki części udziałów w spółce IRPC Polyol na kwotę ok. 80 mln BAHT, co stanowi równowartość ok. 8,9 mln PLN. Ostateczna cena zostanie ustalona na podstawie rocznego sprawozdania finansowego spółki IRPC Polyol za rok 2016, zbadanego przez biegłego rewidenta. (kk)

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